

Dynamical correction to Kohn-Sham conductances from static density functional theory

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For molecules weakly coupled to leads the *exact* Kohn-Sham (KS) conductance can be orders of magnitude larger than the true conductance due to the lack of *dynamical* exchange-correlation (xc) corrections. In this work we show how to incorporate dynamical effects in KS transport calculations. The only quantity needed is the *static* xc potential in the molecular junction. Our scheme provides a comprehensive description of Coulomb blockade without breaking the spin symmetry. This is explicitly demonstrated in single-wall nanotubes where the corrected conductance is in good agreement with experimental data whereas the KS conductance fails dramatically.

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The active field of molecular electronics [1] remains a challenge for *ab initio* methods. Density Functional Theory (DFT) is at present the only viable route for an atomistic description of complex molecular junctions. However, DFT transport calculations still suffer from some conceptual difficulties. Especially thorny is the problem of weakly coupled molecules where level alignment and charging effects play a prominent role. Toher et al. [2] showed that the discontinuity of the exchange-correlation (xc) potential [3] is crucial for opening a gap in the I - V characteristics of closed-shell molecules, and hence for capturing the Coulomb blockade (CB) effect at even electron numbers N . At the same time, the current “DFT understanding” of CB in open-shell molecules is rather limited: while the discontinuity suppresses the DFT conductance at even N , it has the opposite effect at odd N , see below. In this work we provide a comprehensive picture of CB, valid for all N without breaking the spin symmetry. The key ingredient is the *dynamical* xc correction to the conductance [4, 5] which, remarkably, can be expressed exclusively in terms of *static* DFT quantities. Comparison with experiments on single-wall nanotubes shows a significant improvement of the corrected conductances over the DFT results.

At zero temperature and for single-channel junctions the equality $G_s = G$ between the Kohn-Sham (KS) conductance G_s and the true conductance G is a consequence of the Friedel sum rule [6]. The key ingredient to reproduce the Kondo plateau in G_s is the discontinuity [7–9]. However, at temperatures higher than the Kondo temperature T_K , G_s can be orders of magnitude larger than G [7]. We can understand this discrepancy by modeling the molecule as a single level (HOMO or LUMO) of energy v and Coulomb repulsion U coupled to left (L) and right (R) featureless leads contributing $\gamma = \gamma_L + \gamma_R$ to the

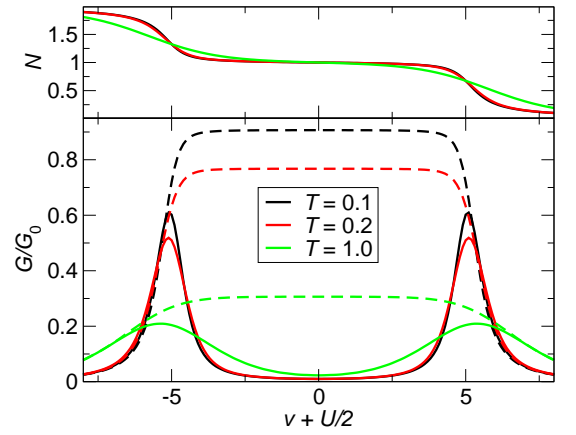


FIG. 1. Top: Electron number N versus gate in MB and DFT (indistinguishable) for a single level coupled to featureless leads with $U = 10$, $\mu = 0$ at various temperatures T (all energies in units of γ). Bottom: G from Eq. (2) (solid) and G_s from Eq. (4) (dashed) in units of $G_0 = 2e^2/h$.

broadening of the spectral peaks. Given the Many-Body (MB) spectral function $A(\omega)$ the number of electrons is

$$N = 2 \int f(\omega) A(\omega), \quad \int \equiv \int \frac{d\omega}{2\pi} \quad (1)$$

whereas the zero-bias conductance reads

$$G = -2 \frac{\gamma_L \gamma_R}{\gamma} \int f'(\omega) A(\omega) \quad (2)$$

with the Fermi function $f(\omega) = 1/(e^{\beta(\omega - \mu)} + 1)$ at inverse temperature $\beta = 1/T$ and chemical potential μ . For temperatures $T \gg T_K$ the Abrikosov-Suhl (AS) resonance is strongly suppressed and the spectral function

is well represented by [10]

$$A(\omega) = n L_\gamma(\omega - v - U) + (1 - n) L_\gamma(\omega - v) \quad (3)$$

where $n = N/2$ and $L_\gamma(\omega) = \gamma/(\omega^2 + \gamma^2/4)$. Consider now the KS system whose spectral function is $A_s(\omega) = L_\gamma(\omega - v - v_{\text{Hxc}}[N])$. The Hartree-xc (Hxc) potential v_{Hxc} is such that the number of electrons N which solves $N = 2 \int f(\omega) A_s(\omega)$ is the same as in Eq. (1). We obtain v_{Hxc} by reverse engineering. In Fig. 1 (top) we show the MB and DFT N - v curves (which are indistinguishable) for three different temperatures. The values of N are essentially independent of T up to $T \sim \gamma$. Having v_{Hxc} we calculate the KS conductance from

$$G_s = -2 \frac{\gamma_L \gamma_R}{\gamma} \int f'(\omega) A_s(\omega). \quad (4)$$

In Fig. 1 (bottom) we compare G and G_s . Despite the fact that the MB and DFT densities are *identical* there is no signature of the CB peaks in G_s . Therefore any DFT approach, based either on nonequilibrium Green's functions or on scattering formalisms, considerably overestimates the conductance of open-shell molecules. We remark that the physical situation discussed here is distinct from that of Ref. 2 where the discontinuity keeps the HOMO doubly occupied and the LUMO empty when gating the molecule (closed shell).

Dynamical xc effects: Open-shell molecules in the CB regime are probably the most striking example of the failure of static DFT methods. We now derive an exact formula for G in terms of time-dependent (TD) DFT quantities [11]. We take the leads as two jellia (the argument can be generalized to more realistic leads) and choose z as the longitudinal coordinate so that $z \rightarrow -\infty$ is in the left lead, $\alpha = L$, whereas $z \rightarrow \infty$ is in the right lead, $\alpha = R$. Let δV^α be the variation in the potential of lead α . This perturbation generates a current [4, 5]

$$\delta I = (\delta V^R - \delta V^L + \delta V_{\text{Hxc}}^R - \delta V_{\text{Hxc}}^L) G_s \quad (5)$$

where $\delta V_{\text{Hxc}}^\alpha = \lim_{t \rightarrow \infty} \lim_{z \rightarrow s_\alpha \infty} \delta v_{\text{Hxc}}(\mathbf{r}, t)$, $s_{R/L} = \pm$, is the asymptotic value of the variation of the Hxc potential δv_{Hxc} . From linear-response TDDFT

$$\delta V_{\text{Hxc}}^\alpha = \lim_{t \rightarrow \infty} \lim_{z \rightarrow s_\alpha \infty} \int d\mathbf{r}' dt' f_{\text{Hxc}}(\mathbf{r}, \mathbf{r}'; t - t') \delta n(\mathbf{r}', t') \quad (6)$$

where f_{Hxc} is the TDDFT kernel and $\delta n(\mathbf{r}, t)$ is the density variation. The assumption of a steady state implies that $f_{\text{Hxc}} \rightarrow 0$ for $|t - t'| \rightarrow \infty$ and $\delta n(\mathbf{r}, t \rightarrow \infty) = s_\alpha \delta n$ for \mathbf{r} in lead α . In Eq. (6) the contribution of the molecular region to the spatial integral is negligible in the thermodynamic limit. If we define $f_{\text{Hxc}}^{\alpha\beta} = \lim_{t \rightarrow \infty} \lim_{z \rightarrow s_\alpha \infty} \int_{\text{lead } \beta} d\mathbf{r}' f_{\text{Hxc}}(\mathbf{r}, \mathbf{r}'; t)$ then $\delta V_{\text{Hxc}}^\alpha = \sum_{\beta=L,R} f_{\text{Hxc}}^{\alpha\beta} s_\beta \delta n$. We emphasize that $f_{\text{Hxc}}^{\alpha\beta}$ is not the static DFT kernel since the limit $t \rightarrow \infty$ is taken *after* the limit $|z| \rightarrow \infty$ and these two limits, in general,

do not commute [12]. This implies that we cannot model $f_{\text{Hxc}}^{\alpha\beta}$ by performing DFT calculations on leads of finite length. Inserting the expression for $\delta V_{\text{Hxc}}^\alpha$ into Eq. (5) we find $\delta I = (\delta V^L - \delta V^R) G_s - \Phi G_s \delta n$ where

$$\Phi \equiv f_{\text{Hxc}}^{RL} + f_{\text{Hxc}}^{LR} - f_{\text{Hxc}}^{RR} - f_{\text{Hxc}}^{LL}. \quad (7)$$

The expression for δI is correctly gauge invariant. The kernel f_{Hxc} is defined up to the addition of an arbitrary function $g(\mathbf{r}) + g(\mathbf{r}')$ [13] and we see that Φ is invariant under this transformation. In conclusion

$$G \equiv \frac{\delta I}{(\delta V^R - \delta V^L)} = \frac{G_s}{1 + \chi \Phi G_s}. \quad (8)$$

The quantity $\chi \equiv \delta n / \delta I \sim 1/(v_F \sigma)$ with v_F the Fermi velocity and σ the cross section of the leads [14]. Thus all dynamical xc effects are contained in Φ .

Approximations to Φ : By definition the product $\chi \Phi = (\delta V_{\text{Hxc}}^R - \delta V_{\text{Hxc}}^L) / (\delta V^R - \delta V^L)$ is the ratio between the Hxc bias and the physical bias. To gain some insight into its density dependence we consider again the single level model. For $N \neq 1$ the real and KS systems respond similarly and consequently $G \sim G_s$. On the other hand for $N = 1$ we have $G \sim 0$ whereas $G_s \sim G_0 = 2e^2/h$ the quantum of conductance. Therefore Φ is small for $N \neq 1$ and large for $N = 1$. Interestingly the quantity $\partial v_{\text{Hxc}} / \partial N$ behaves similarly. Is there any relation between Φ and $\partial v_{\text{Hxc}} / \partial N$? If so this relation would simplify enormously the problem of estimating the dynamical xc correction since $\partial v_{\text{Hxc}} / \partial N$ can be calculated from static DFT. In the following we show that in the CB regime this relation does actually exist.

Consider the system in equilibrium. Using Eq. (1) the compressibility $\kappa = \partial N / \partial \mu$ can be written as $\kappa = \frac{\gamma}{\gamma_L \gamma_R} G + 2 \int f(\omega) \frac{\partial A(\omega)}{\partial \mu}$, where we identified the conductance G of Eq. (2). If we define the quantity $R \equiv -2 \int f(\omega) \frac{\partial A(\omega)}{\partial N}$ then $\kappa = \frac{\gamma}{\gamma_L \gamma_R} G \frac{1}{1+R}$. As the MB and DFT densities are the same, the MB and DFT compressibilities are the same too. Hence $\kappa = \frac{\gamma}{\gamma_L \gamma_R} G_s + 2 \int f(\omega) \frac{\partial A_s(\omega)}{\partial \mu}$, where we identified the KS conductance G_s of Eq. (4). The KS spectral function depends on μ through N , and the dependence on N is all contained in v_{Hxc} : $\frac{\partial A_s}{\partial \mu} = -\frac{\partial A_s}{\partial \omega} \frac{\partial v_{\text{Hxc}}}{\partial N} \frac{\partial N}{\partial \mu}$. Using this result under the integral sign, solving for κ and equating the MB and DFT expressions we get

$$\frac{G}{G_s} = \frac{1 + R}{1 + \frac{\gamma}{\gamma_L \gamma_R} G_s \frac{\partial v_{\text{Hxc}}}{\partial N}}. \quad (9)$$

No approximations have been made so far. Let us study the dependence of R on temperature.

We first consider the low temperature case. For simplicity we take $\gamma_L = \gamma_R$ and set $v = -U/2$ at the particle-hole (ph) symmetric point. At zero temperature $G = G_s = G_0$ and hence $R = \frac{4G_0}{\gamma} \frac{\partial v_{\text{Hxc}}}{\partial N}$. The correctness of this equality can easily be verified. The

compressibility is weakly dependent on temperature up to $T \sim \gamma$ [15]. Therefore the v_{Hxc} obtained by reverse engineering is a good approximation even for $T = 0$. At the ph symmetric point ($N = 1$) this approximation gives $\partial v_{\text{Hxc}}/\partial N \sim U^2/\gamma$ [16]. At $T = 0$ the MB spectral function for $\omega \sim \mu$ is dominated by the AS resonance $A(\omega \sim \mu) \sim \frac{4T_K}{\gamma} L_{T_K}(\omega - \mu)$ and thus $\int f(\omega) \frac{\partial A(\omega)}{\partial \mu} \sim -\frac{2}{\pi\gamma}$. For the compressibility we have from the Bethe-Ansatz [18] $\kappa = (8\gamma)/(\pi U^2) [1 + \mathcal{O}(\gamma/U)]$ from which it follows that also R goes like $(U/\gamma)^2$. For temperatures $T > T_K$ the AS resonance broadens and its height decreases like $h(T/T_K)$ where h is a universal function which approaches zero at high T [17]. This means that $R \sim h(T/T_K)(U/\gamma)^2$ remains large until the AS resonance disappears. No simple relation between Φ and $\partial v_{\text{Hxc}}/\partial N$ exists when Kondo correlations are present.

At temperatures $T \gg T_K$ thermal fluctuations destroy the Kondo effect and the MB spectral function is well approximated by Eq. (3). Therefore $R(v) = I(v) - I(v+U)$ where $I(E) \equiv \int f(\omega) L_\gamma(\omega - E)$. Hence $R(v) \sim 0$ for $v > \mu$ ($N \sim 0$) or $v < \mu - U$ ($N \sim 2$) and $R(v) \leq 1$ otherwise. We can derive a more convenient expression for R by inserting Eq. (3) into Eq. (1) to find $N = N I(v+U) + (2-N) I(v)$ and hence $1 + R = 2I(v)/N$. Unfortunately $I(v)$ is not an explicit “density functional” due to the implicit dependence of $v = v[N]$. However, for $N < 1$ a good approximation to N is $N \sim 2I(v)/(1 + I(v))$ which implies $I(v) = N/(2-N)$. Taking into account the ph symmetry we can approximate R by an explicit density functional as $1 + R \sim 2/(1 + |\delta N|)$ where $\delta N = N - 1$. Inserting this into Eq. (9) we deduce our main result

$$\frac{G}{G_s} = \frac{2}{1 + |\delta N|} \frac{1}{1 + \frac{\gamma}{\gamma_L \gamma_R} G_s \frac{\partial v_{\text{Hxc}}}{\partial N}} \quad (10)$$

which provides a simple correction to the KS conductance. Remarkably, the dynamical xc correction of Eq. (8) is *entirely* expressed in terms of *static* DFT quantities of the molecular junction. In Fig. 2 we compare the conductance G of Eq. (2) with the conductance calculated from Eq. (10). Even though the approximate R is not on top of the exact one, see inset, the agreement between the two conductances is extremely good. The position, width and height of the peaks as well as the decay for large $|v|$ are all well reproduced. In fact, the approximate R is rather accurate except in the region where $N \sim 1$ ($\mu - U < v < \mu$). In this region, however, $\partial v_{\text{Hxc}}/\partial N$ is large and the small error in R does not significantly affect G .

Application to physical systems: In real molecules v_{Hxc} is a \mathbf{r} -dependent functional of the density. We write $v_{\text{Hxc}}(\mathbf{r}) = \delta v_{\text{Hxc}}(\mathbf{r}) + \bar{v}_{\text{Hxc}}$ as the sum of a functional δv_{Hxc} with a weak dependence on $N = \int_V d\mathbf{r} n(\mathbf{r})$ and a spatially uniform part $\bar{v}_{\text{Hxc}} = \frac{1}{V} \int_V d\mathbf{r} v_{\text{Hxc}}(\mathbf{r})$, where the integral is over the volume V of the molec-

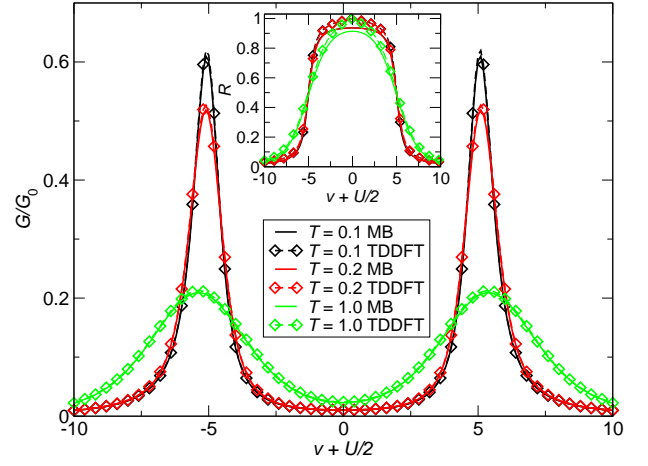


FIG. 2. Conductance from Eq. (2) (solid) and from Eq. (10) (dashed). The inset shows a comparison between the exact and the approximate R . Same parameters as in Fig. 1.

ular junction. For weakly coupled molecules \bar{v}_{Hxc} exhibits sharp steps as a function N when N crosses an integer. If we are at resonance and spin fluctuations are suppressed (no Kondo effect) then the KS conductance should be corrected according to Eq. (10) in which $\partial v_{\text{Hxc}}/\partial N \rightarrow \partial \bar{v}_{\text{xc}}/\partial N$. Below we argue that this correction applies out-of-resonance too. Let μ be in the HOMO-LUMO gap and consider a two-level system with Γ_α the 2×2 broadening matrix. For general Γ_α no simple analytic relation between G and N exists. However if $\Gamma_{\alpha,ml} = (\gamma_\alpha/2)\delta_{ml}$ then $N = 2 \int f(\omega) \text{Tr}[A(\omega)]$ and $G = -2 \frac{\gamma_L \gamma_R}{\gamma} \int f'(\omega) \text{Tr}[A(\omega)]$. Discarding the dependence of δv_{Hxc} on μ (which is weak by definition) we can go through the same steps of the single-level derivation and find again Eq. (10). It is therefore reasonable to expect that the KS conductance should be corrected even out-of-resonance (closed shell) and that this correction should be proportional to $G_s \partial \bar{v}_{\text{Hxc}}/\partial N$.

We propose a scheme to calculate G from DFT. Given the KS Hamiltonian matrix $h_{\text{KS},ml} = \delta_{ml}\epsilon_l$ and the broadening matrices $\Gamma_{\alpha,ml}$ we determine the density and G_s in the usual manner. G is then obtained from Eq. (10) where δN is the deviation of $(N - \text{Int}[N])$ from 1 whereas $\gamma_\alpha = \gamma_\alpha(N) \sim \Gamma_{\alpha,HH}$ if $\mu \sim \epsilon_H$ (resonance, open shell) and $\gamma_\alpha(N) \sim \frac{1}{2}(\Gamma_{\alpha,HH} + \Gamma_{\alpha,LL})$ if $\mu \sim \frac{1}{2}(\epsilon_L + \epsilon_H)$ is in the HOMO-LUMO gap (out-of-resonance, closed shell). One could improve the approximation to γ_α using different weights, but the qualitative features of the results are independent of these details, see below.

To illustrate the importance of the dynamical xc correction we consider two paradigmatic junctions in which δv_{Hxc} can be discarded. For \bar{v}_{Hxc} we choose a best fit of the zero-temperature limit of the single-level Hxc potential, but now sum over all possible charged states of the

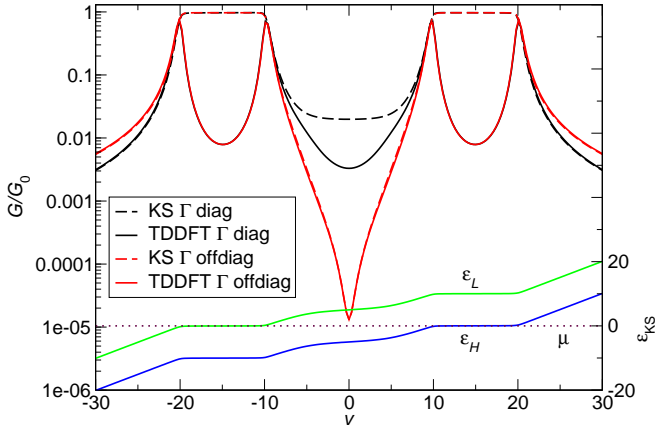


FIG. 3. KS conductance (dashed) and TDDFT conductance from Eq. (10) (solid) for the HOMO-LUMO model with diagonal and off-diagonal Γ -matrices (left axis) and KS energies $\epsilon_{H/L} = \epsilon_{0H/L} + \bar{v}_{\text{Hxc}}$ (right axis).

molecule [19], i.e.,

$$\bar{v}_{\text{Hxc}} = \sum_K \frac{U(K)}{\pi} \arctan\left(\frac{N-K}{W(K)}\right). \quad (11)$$

The charging energies $U(N)$ are given by the xc part of the derivative discontinuity of the molecule with N electrons [3]. For the widths we take $W(N) = 0.16 \gamma(N)/U(N)$ which is consistent with Ref. 16

HOMO-LUMO model: We consider a two-level system which has 2 electrons in the HOMO in the charge neutral state. Let $\epsilon_{0H} = -\epsilon_{0L} = -\epsilon_0 < 0$ be the noninteracting single-particle energies, $\Gamma_L = \Gamma_R = \frac{\gamma}{2} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix}$ and $U(N) = U$ independent of N . We solve the self-consistent equation for the density with $U = 10$, $\epsilon_0 = 5$, $\mu = 0$ and $\beta = 10$ (all energies in units of γ). G_s and G from Eq. (10) are shown in Fig. 3 (left axis). As expected the discontinuity of v_{Hxc} opens a gap in G_s for even N , in agreement with the results of Ref. 2. Here the dynamical xc correction only weakly affects G_s since $\partial v_{\text{Hxc}}/\partial N$ is multiplied by $G_s \ll 1$. On the other hand for odd N the KS conductance exhibits a Kondo plateau due to the pinning of the KS level to μ , see right axis. This is the regime previously discussed and no CB is observed. The dynamical xc correction remedies this deficiency. We remark that the results remain essentially unaltered if the off-diagonal matrix elements of Γ_α are discarded which amounts to neglecting interference effects [20].

SWNT: Experimental evidence of CB oscillations has been recently reported in metallic single-wall nanotubes (SWNT) quantum dots [21, 22]. The finite length of the SWNT causes a level quantization of the twofold degenerate bands. Following Ref. 21 we model the noninteracting single-particle energy levels as $\epsilon_{0l\nu} = l\Delta + l(\nu-1)\delta$ where $\nu = 0, 1$ is the band index and l is the integer of

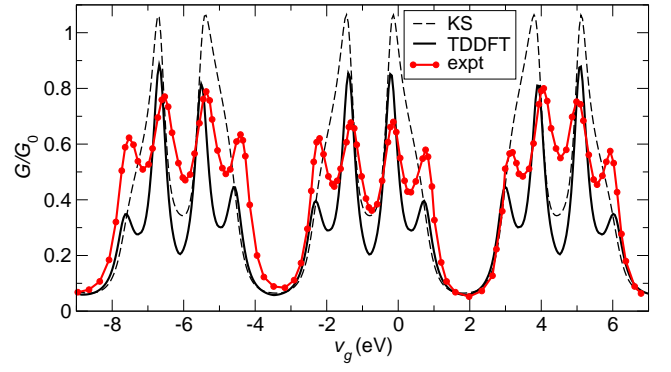


FIG. 4. KS and TDDFT conductance (Eq. (10)) for a SWNT quantum dot in comparison to experimental conductance from Ref. 21, as function of gate voltage v_g .

the quantized wavevector along the nanotube axis. Since the wavevector is a good quantum number our approximation $\delta v_{\text{Hxc}} = 0$ is justified. The constant interaction model [23] has been refined by Oreg *et al.* [24] to account for the observed fourfold periodicity in the electron addition energy. We choose the $U(K)$'s in Eq. (11) according to [25]: $U(1) = U(3) = E_C + \delta U + J$, and $U(2) = U(4) = E_C - \delta U$, and, for $K > 4$, $U(K) = U(i)$ if $K - i = 0 \pmod{4}$ with $i = 1, 2, 3, 4$. The average values of these parameters can be found in Ref. 21. Here we change them slightly to match the peak positions of the SWNT of length ~ 100 nm (all energies are in meV): $\Delta = 9.2$, $\delta = 2.27$, charging energy $E_C = 2.485$, exchange energy $J = 0.7$, extra charging energy for doubly occupied levels $\delta U = 0.37$. For the broadening matrix we make the approximation $\Gamma_{\alpha,ml} = (\gamma/2)\delta_{ml}$ (no visible interference from the experiment). In Fig. 4 we compare the KS, TDDFT and experimental conductance versus the gate voltage $v_g = v_0 + \alpha v$ where v_0 is chosen in order to have the same reference energy as in Ref. 21 and we estimated the ratio $\alpha = C/C_g \sim 250$ between the total capacitance and the gate capacitance from the experimental data. In our calculation we have taken into account 22 single-particle levels and chose a temperature $T \ll \gamma \sim 1$ meV. Despite the simplicity of the model we clearly see that the dynamical xc correction improves considerably over G_s which exhibits two deformed Kondo plateaus per period and misses the fourfold periodicity. The qualitative behavior of G and G_s does not change by varying the parameters within a reasonable range around the average values reported in Ref. 21 (not shown).

In conclusion we highlighted the importance of the discontinuity for an accurate G_s and for an accurate dynamical xc correction to G . Approaches to generate discontinuous xc potentials are emerging both in the static [26] and dynamical [27] case. Our scheme provides a coherent picture of CB within (TD)DFT without breaking the spin symmetry. By application to two different model

molecular junctions we showed that the dynamical xc correction reduces G_s thus contributing to close the gap between theory and experiments.

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